Development of Optical Systems for Hard X-rays at SPring 8 Yoshio Suzuki JASRI/SPring-8

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Development of Hard X-ray Optics at SPring-8

- 1. Beamline configurations,
- Fresnel zone plate optics
 Micro- & nano-beam generation and characterization,
 Imaging microscopy and tomography applications,
 Holography and phase-contrast imaging,
 Use of quasi-monochromatic (direct undulator) radiation,
- 3. Total reflection mirror optics,
- 4. Sputtered-sliced zone plate optics,
- 5. Refractive lens optics,
- 6. Theory of resolution limit.



Overview of Beamlines

BL20XU (Coherent optics, Microbeam, Holography, Interferometer, etc.) Undulator, Medium Length (248 m) beamline.
BL20B2 (Computer Tomography, Topography, Medical Imaging, etc.) Bending Magnet, Medium Length (215 m).
BL37XU (Fluorescent X-ray Micro-analysis) Undulator, Normal Length (50 m) beamline.
BL47XU (Microbeam, Imaging Microscopy & Micro-CT)

Undulator, Normal Length (50 m) beamline.

(BL28B2: BM white beam, topography, BL29XU: RIKEN, 1 km BL)

Energy range:

5-113 keV,

Spatial Coherence:

~1 mm @ 1Å (BL20XU),

Beam Cross-section:

~ 30 nm (micro-focus) - 300 mm (215 m-beamline with BM source). Flux Density:

~10E14 photons/s/mm2 (direct beam),

> 10E11 photons/s/ μ m2 (micro-focus).



248 m-long Beamline, BL20XU, at SPring-8



Experimental setup

Application of Coherent X-ray Beam -X-ray In-line Holography -



Measured Point-spread-function

"Zooming Tube"
Hamamatsu Photonics C5333
Photo-cathode: CsI (~2000Å),
Magnification: 5-240,
Resolution (point-spread-function): 0.7 μm in FWHM.



EX-ray Energy: 8 keV

X-ray Energy: 80 keV

Object: Gold Wire, 50 μ m Diameter, Imaging Detector: Zooming Tube, C5333, Hamamatsu Photonics, Field of View: 300 μ m in Diameter.

Measured Hologram of Test Object

Measurement of X-ray Coherence using Two-beam Interferometer with Prism Optics



Schematic diagram of experimental setup at beamline 20XU of SPring-8



Optical system for two-beam interferometer with X-ray prism.



Typical interference fringe patterns measured at an X-ray wavelength of 1 Å. Beam deflection angle: $\Delta \theta = 44 \ \mu rad$. Slit dimensions: 18 $\mu m \ge 19 \ \mu m$. Measured fringe spacing is 2.3 μm . Beam overlap is 300 μm . Exposure time is 60 s.



(a) 1.5 Å and 3 degrees (b) 1.0 Å and 2 degrees (c) 0.5 Å and 1 degree

Interference Fringes measured with X-ray Zooming Tube.

Prism to detector distance: 6.9 m, Field of view : 100 μ m in diameter, X-ray wavelength: 0.5, 1.0, and 1.5 Å, Glancing angle to prism surface: 1.0, 2.0, and 3.0 degrees.

Cu grid mesh 64 μm pitch



Test Patterns 5 μm L/S

Hologram

Reconstructed Image

Leith-Upatnieks Type Two-beam Holography $\lambda = 1.0$ Å, Sample to image detector: 6.7 m



Visibility of interference fringes. Solid squares, circles and triangles represent experimental data, Solid line, dotted line and dashed line are respective theoretical curves for source size of 100 μ m, 50 μ m, and 20 μ m. **Development of Hard X-ray Optics at SPring-8**

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Experimental Setup of X-ray Microbeam/Scanning Microscopy at BL20XU **Specification of Fresnel zone plate**

Diameter: 150 μm, Designed focal Length: 100 mm at 8 keV,
 Outermost zone width (d_N): 0.1 μm.
 Diffraction limit (=1.22d_N): 0.12 μm, numerical aperture: 7.5 x 10⁻⁴ at 8 keV,
 Zone material: Ta, 1 μm-thick,
 Supporting membrane: Si₃N₄, or SiC, 2 μm-thick.
 Fabrication method: electron-beam lithography technique at NTT-AT



Schematic Drawing of Zone Plate Structure



Focused Beam Profile Measured by Knife-edge Scan

FZP: Ta 1 μ m-thick, Outermost Zone Width: 0.1 μ m, EB-lithography at NTT-AT, Focal Length: 100 mm @8 keV.



Diffraction Efficiency of Ta-FZP

Closed Circle: Experimental Results,

Solid Line: Calculated Efficiency assuming the Thickness of 1μ m.

Total flux of microbeam: 10^9 photons/s, Focused beam size: $0.12 \ \mu$ m.



Dark-field Knife-edge Scan Method for Focus Test

Primary beam (direct beam) is cut off with an aperture in front of X-ray detector. The scattered X-rays are selected by the aperture. **Advantages of dark-field method:**

- 1. Thin and phase object as knife-edge,
- 2. No differential processing,
- **3. Precise measurement.**

• Result of Numerical Simulation



Simulation of Dark-field Knife-edge Test







Resolution Test of Linear Translation Stage

Kohzu Seiki, type YA-05-14. Stepping Motor, Oriental Motor PX535MH-B. Motor Driver, Melec, micro-step drive H-583. Position Monitor, KEYENCE LC2420.



Test Result of Linear Translation Stage

Kohzu Seiki, type YA-05-14. Stepping Motor, Oriental Motor PX535MH-B. Motor Driver, Melec, micro-step drive H-583. Position Monitor, KEYENCE LC2420. Lifetime of Ta-FZP for radiation damage ~ 3 days in air, >> 1 month in vacuum or in He.





FZP: 100 μ m diameter, 0.25 μ m outermost zone width, X-ray energy: 10 keV @BL47XU, Flux density: ~ 5 x 10¹³ photons/s/mm, Total flux: ~ 10¹⁷ photons

Damage problem is solved at present!



Schematic Diagram Experimental Setup for Imaging Microscopy and Micro-tomography

@ BL47XU



Imaging Microscopy

Objective & Sample: FZP with 0.25 μ m outermost zone width, X-ray Energy: 8 keV.





- Coherent Illumination -
- Incoherent Illumination-

Imaging Microscopy with FZP Objective - Effect of Beam Diffuser -

X-ray energy: 8 keV

Control of coherence is important!



Stony Meteorite Allende
8 keV, x7.61, BM3(x10), voxel size 0.13 μm.
100 projection, exposure time:15 s/projection.



Diatom "Achnanthidium lanceolata" 8 keV, x10, BM3(x10), voxel size 0.1 μ m. 360 projection, exposure time: 60 s/projection.

X-ray Micro-tomography using Imaging Optics with Fresnel Zone Plate Objective

Low-emittance SR source is not suitable for imaging microscopy, because of

high spatial coherence: Small source size (~ 10 μ m vertical x 100 μ m horizontal), Small divergent angle (~ 10 μ rad).

Critical illumination with simple condenser lens: F-number matching --> small field of view (< a few μm), Coherent illumination --> Speckle noise.

Critical illumination and Köhler illumination (best optics for imaging microscopy)

Critical illumination:

Demagnified image of source at the object plane., Each point of source corresponds to each point of field of view, Not suitable for 3rd generation SR source.

Köhler's illumination:

Infinite focus, Each points at source to each angle of illuminating beam.

Illumination Optics for Imaging Microscopy

First experiment on imaging microscopy at SPring-8:

Parallel beam illumination

--> Edge-enhancement artifact, Strong speckle noise.

2nd Step:

Partial Coherent illumination by diffuser

--> Less artifacts, and no speckles, Weak edge-enhancement, Nonuniform imaging properties in the field of view, Asymmetric feature of imaging properties. (off-axial illumination)

Need of Condenser Optics for Imaging microscopy.



$$\Delta = 0.61 \ \lambda/NA,$$

NA: numerical aperture of objective lens.

 $NA = \sin\theta$.

Useful formula:

$$\Delta = 1.22 \ dr_{\rm N}$$

 $dr_{\rm N}$: Outermost zone width

Parallel beam illumination: $\Delta = 0.82 \lambda/NA$,

With condenser optics of 1.5NA: $\Delta = 0.57 \lambda/NA$,

Example: dr_N = 100 nm, Δ = 122 nm. NA = 7.75 x 10⁻⁴ at 8 keV. (8 keV = 1.55 Å)

Spatial Resolution of FZP Microscope



Diffraction-limited resolution = Geometrical defocusing

 $2D \ge NA = 0.61 \lambda/NA$,

Depth of focus (tolerance of sample thickness):

 $2D = 0.61 \lambda / NA2.$

In tomography measurement, Depth of focus > Sample diameter. Example: $\Delta = 122 \text{ nm} (dr_N = 100 \text{ nm}),$ $NA = 7.75 \text{ x } 10^{-4} \text{ at } 8 \text{ keV}.$

 $2D = 157 \ \mu m.$

Depth of Focus



Second order approximation and the Rayleigh's quarter wavelength rule:

$$\begin{aligned} &|\{-r_{\rm a} r_{\rm n} \cos \phi/a + r_{\rm b} r_{\rm n} \cos \phi/b\} + 1/2 \{ r_{\rm n}^{2}/a + r_{\rm n}^{2}/b - n\lambda \} \\ &- 1/8 \{ (r_{\rm a}^{2} - 2r_{\rm a} r_{\rm n} \cos \phi + r_{\rm n}^{2})^{2}/a^{3} - r_{\rm a}^{4}/a^{3} + (r_{\rm b}^{2} + 2r_{\rm b}r_{\rm n} \cos \phi + r_{\rm n}^{2})^{2}/b^{3} - r_{\rm b}^{4}/b^{3} \} | < \lambda/4. \end{aligned}$$

Aberration Theory of FZP Microscope by Wave Optics

Depth of focus (tolerance of sample thickness for tomography):

$$2D = 0.61 \ \lambda/NA^2,$$

 $NA \sim r_N/f, \ D \sim r_a \quad ---> \quad r_a r_N^2/f^2 < 0.3\lambda$
 $r_N/f << 1, \ r_a/f << 1 \text{ for hard -X-ray FZP.}$

Other wavefront aberrations:

 $3r_{a}^{2}r_{N}^{2}/f^{3} < \lambda,$ $2r_{a}r_{N}^{3}/f^{3} < \lambda,$ $1/2r_{N}^{4}/f^{3} < \lambda$

Chromatic aberration: $\Delta\lambda/\lambda < 0.61/N$, (*N*: total zone number).

 $\Delta\lambda/\lambda \sim 10^{-4}$ (crystal monochromator, Si 111)

Example: f = 100 mm at 8 keV, $r_{\rm N} = 77.5 \ \mu \text{m},$ N = 388, $NA = 7.75 \ \text{x} \ 10^{-4} \text{ at 8 keV}.$ $(\lambda = 1.55 \ \text{\AA})$ $M \sim 70$

Most serious aberration is Depth of Focus!



CT image of IDP (L2008D3 #17). (a) three dimensional reconstruction from CT images. (b) sagital slice derived from three dimensional reconstructed image.

Hard X-ray Imaging Micro-tomography

Fresnel zone plate = Chromatic aberration,

Requirement on monochromaticity for Fresnel zone plate ~ Number of Fresnel zone.

---> $\Delta\lambda/\lambda$ < 1/N (number of Fresnel zone)

 $N \sim or > 100$, (requirement for natural lens approximation).

 $\Delta\lambda/\lambda \sim 10^{-4}$ with crystal monochromator, too narrow! ---> loss of photon flux.

Use of direct undulator radiation, Δλ/λ ~ 100.
High flux microbeam, Short Exposure Time. **BL40XU of SPring-8 (High Flux Beamline)**

1. Undulator radiation without monochromator, $\Delta\lambda/\lambda \sim 1.2\%$ @ $\epsilon = 3 \text{ nm rad}$

Helical Undulator --> Suppression of higher order,
 Condenser Optics: K-B mirror



Measured Spectra of Undulator Radiation Front-end Slit Aperture: 15 μrad (horizontally) x 5 μrad (vertically)

Available flux ~ 100 times that at conventional beamlines (undulator beamlines with crystal monochromator.

X-ray Microbeam & Imaging Microscopy with Sub-micron resolution and high flux! (~ 100 times, compared with conventional beamlines)



Experimental Setup for Imaging Microscopy at BL40XU SPring-8



<u>10 µm</u>

Object: Cu mesh, 2000 lines/inch Object: Fresnel zone plate, 0.25 μm outermost zone width

Image of test object

Objective: FZP, 0.25 μ m outermost zone width, 100 zones, Magnification: 11.3, X-ray energy: 8.34 keV, Exposure time: 1.5 ms (Single Shot)

Hard X-ray Imaging Microscopy with Fresnel Zone Plate Objective & Quasi-monochromatic Undulator Radiation at BL40XU



Optical Layout of Microbeam Experiment at BL40XU of SPring-8



Measured Profiles of Focused Beam

X-ray Energy: 8.317 keV Total Flux of Focused Beam: ~ 2 x 10¹² photons/s



151 x 334 pixels,
0.3 μm/pixel,
0.3 s dwell time.

66 x 126 pixels,
0.2 μm/pixel,
0.2 s dwell time.

Scanning Microscopic Images of Resolution Test Patterns

Microbeam and Scanning Microscopy with FZP and Quasi-monochromatic Undulator Radiation



Experimental Setup of X-ray Microbeam/Scanning Microscopy with Total-reflection Mirror Optics (Kirkpatrick-Baez Configuration)

Kirkpatrick-Baez Optics with Aspherical (Plane Parabola) Mirrors, L1: 45 mm, L2: 45 mm, L3: 25 mm, *f*: 75 mm, Glancing angle: 2.8 mrad. (Pt coated SiO₂), Fabricated at Cannon Co. Japan.



Micirobeam and Scanning Microscopy with Total-reflection Mirror Optics



- Diffraction limit
- --- Geometrical Size



Energy Dependence of Resolution



Reflectivity of Total Reflection Mirrors

Pt surface, Glancing angle: 2.8 mrad.



Total Reflection Mirror for High Energy X-ray Microbeam



Total Reflection Mirror for High Energy X-ray Microbeam



Schematic View of Sputtered-sliced Zone Plate



Fabrication Process of Sputtered-sliced Fresnel Zone Plates



SEM Image of Sputtered-sliced Fresnel Zone Plate

Au Core (50 μ m in diameter), Cu/Al 50 Layers, Outermost zone width of 0.15 μ m.



Diffraction efficiency: 25% @ 1.4 Å Sagittal Focus

Sagittal Focus (1/4 of annular aperture)

Focused Beam Profile Measured by Edge-scan @BL20XU









0.2 μ m line & space

X-ray wavelength: 1.4 Å, 128 x 64 pixels, 0.0625 μm/pixel, Dwell time: 0.4 s/pixel.



0.1 μ m line & space

X-ray wavelength: 1.0 Å, 256 x 70 pixel, 0.0625 μm/pixels, Dwell time: 0.4 s/pixel.

Scanning Microscopic Image of Resolution Test Pattern



X-ray Energy: 82 keV (0.151 Å), f ~ 700 mm, Cu/Al sputtered-sliced FZP (50 layers), Core (center beam stop): Au 50 μ m in diameter, Outermost zone width: 0.15 μ m, Thickness: ~ 40 μ m.





Scanning Microscopic Image

Sample: gold mesh (1500 lines/inch), X-ray Energy: 82 keV ' 51 x 51 pixels, 1 μm/pixel, Dwell time: 2 s/pixel, CdZnTe-detector for fluorescent X-rays.

Microfocusing/scanning microscopy with SS-FZP at 82 keV

Diffraction efficiency: 15%



Microbeam with Sputtered-sliced FZP

Focused Beam Profile Measured by Edge-scan @BL20XU

X-ray wavelength: 0.124 Å (100 keV), f ~ 900 mm, Cu/Al sputtered-sliced FZP (70 layers), Core (beam stop): Au 50 μ m in diameter, Outermost zone width: 0.16 μ m, Thickness: ~ 180 μ m.

Resolution Limit of X-ray Microscope

General Theory Rayleigh's criterion (Diffraction Limit in Classical Optics)



NA = n sinθ,
n: Index of Refraction,
C ~ 1 (constant, dependent on optics configuration).

Typically, C ~ 0.61 (Circular aperture), n ~ 1 (in air), $\sin\theta \sim 0.5$ (F ~1) for visible light,

$\delta \sim \lambda$: Resolution limit of Optical Microscope.

General Theory

Uncertainty Principle (Quantum Mechanics) $\Delta p \Delta x \ge h$

Momentum of Photon: h/λ , Momentum Spread by Focusing Optics: $\Delta p = 2|p| \sin\theta$.



How about hard X-ray microscopy?

Total Reflection Mirror Optics



 ρ (g/cm3): Density of Mirror Material, λ (nm): X-ray wavelength.

The theoretical limit of spatial resolution, Δx , is determined only by the density of the reflector surface material, $\sqrt{\rho}$. The limit of spatial resolution is approximately 10 nm.

For combined mirror optics (Wolter-type-mirror or tandem-toroidal-mirror optics),

$$\Delta \mathbf{x} = \mathbf{0.61} \mathbf{x} \,\lambda/(4\theta \mathbf{c}). \tag{4}$$

1. P. Kirkpatrick and A. V. Baez: J. Opt. Soc. Am. 38 (1948) 766.

2. Von H. Wolter: Ann. Physik 10 (1952) 94.

3. Y. Sakayanagi: Optica Acta 23 (1976) 217.

X-ray Wave Guide

Planar wave guide, 1-D solution,

Boundary Condition: $2d \sin\theta = m\lambda$, m = 1, 2, 3,

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Lowest mode of propagating wave: m = 1,
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d sin $\theta = \lambda/2$,



d: gap of waveguide (inner diameter of waveguide) θ: glancing angle to wall

 $\theta \leq \theta c$ (θc : critical angle for total reflection)

When the phase jump at total reflection = π (case of $\theta \ll \theta c$), minimum size of wave guide, do,

do = $\lambda / (2\theta) \le \lambda / (2\theta c)$.

However, the penetration depth of evanescent wave, t,

 $t \sim \lambda/(\theta c^2 - \theta^2)^{1/2}$. So, effective broadening of wavepacket is

 $\Delta x \sim t \sim \lambda/\theta c$: the same as that of total reflection mirror optics.

or $\Delta x \leq \lambda/(2\theta c)$, simply from uncertainty principle.

cf. C. Bergemann, H. Keymeulen and J. F. van der Veen: Phys. Rev. Lett. 91 (2003) 204801.



Electric Field Intensity in the Wave-guide Si ($\rho = 2.34$), $\lambda = 1.28$ Å, d = 60 Å, and d = 160 Å.

Refractive Lens Optics



Refractive Lens: Exact Solution

Diffraction-limited Resolution of Single Refractive Lens

Considering phase shit of $2m\pi$, (m λ , m = 1, 2, 3,)

$$(x^{2} + y^{2})^{1/2} + n(f - x) = f + m\lambda, \qquad (8)$$

 $[x - {f + m\lambda/(1 - n)}n/(1 + n)]^{2}/[{f + m\lambda/(1 - n)}^{2}/(1 + n)^{2}] + y^{2}/[{f + m\lambda/(1 - n)}^{2}(1 - n)/(1 + n)] = 1.$ (9)

Numerical Aperture of the Lens (NA):

NAmax =
$$[(1 - n)/(1 + n)]^{1/2}/[1/(1 + n)] = (1 - n^2)^{1/2}$$
. (10)



Using $n = 1 - \delta$, and $\delta \ll 1$,

cf. Y. Suzuki, Jpn. J. Appl. Phys. 43 (2004) 7311-7314.

Expansion to Fresnel Lens and Fresnel Zone Plate

The nesting configuration, the series of ellipsoids m = 0, 1, 2, 3, ..., MFresnel zone plate at f = x:

$$(f^2 + y^2)^{1/2} = f + m\lambda.$$
 (14)

$$y = [2m\lambda f + (m\lambda)^2]^{1/2}.$$
 (15)

When $f \gg m\lambda$, by neglecting the higher-order terms,

$$\mathbf{y} = (2\mathbf{m}\lambda f)^{1/2}.$$
 [Zone Plate Equation] (16)

The major axis of the ellipse: $\{f + m\lambda/(1 - n)\}/(1 + n)$,

The major axis of the ellipsoid for the outermost zone should be smaller than the focal length *f*.

 ${f + m\lambda/(1 - n)}/(1 + n) \le f.$ (17)

The possible outermost zone for the planar zone plate:

$${f + M\lambda/(1 - n)}/(1 + n) = f.$$

M: the maximum m.

$$M\lambda/(1 - n) = n f.$$

$$\theta$$
max = $(2M\lambda f)^{1/2}/f$
= $[2f^2n(1 - n)]^{1/2}/f$
~ $(2\delta)^{1/2}$,



cf. Y. Suzuki, Jpn. J. Appl. Phys. 43 (2004) 7311-7314.

Theoretical Resolution Limit of Total Reflection Mirror Optics, Wave-Guide, Refractive Lens, Fresnel Zone Plate

 $\sqrt{(2\delta)} \sim 10$ nm in hard X-ray Region

Possible Ways to Nanometer Resolution

1. Combined Refractive Lens,



Diffraction Limited Resolution ~ 0.61 x $\lambda/(N\theta c)$ N: Number of Combined Lens

Spherical lens might be feasible, because smaller lens has smaller aberration.

cf. C. Schroer and B. Lengeler, Phys. Rev. Let. 94 (2005) 054802

Possible Ways to Nanometer Resolution

2. Three Dimensional Zone Plate (Volume Zone Plate or Laue Lens)



with Optical Path Difference of $m\lambda$.

H. C. Kang et al., Phys. Rev. Lett. 96 (2006) 127401, C. Schroer, Phys. Rev. B 74 (2006) 033405.

Ideal only on Focusing Property.

Next limit: atom size with $\lambda/4$ rule (Rayleigh limit) ~ 1 nm.

ERL & FEL

Complementary?

Which is better for users? Time structure & spectral structure.

Nano-optics: Applications? Users? Practical? R&D of optics: 10 nm resolution -> 1 nm resolution..?

Problems in the 3rd-generation SR source, Spring-8 Most of users and experiments are 2nd generation! Important Problems in Coherent X-ray Sources

- 1. Vibration: optics, light source, ground&building
- 2. Temperature stability: ~0.01° environment.
- 3. Radiation damages, cooling. Same as Spring-8?
- 4. Speckles:
 - No optics without any speckles.
- 5. No optics is best optics?